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Comparison of pollutant emission control strategies for cadmium and mercury in urban water systems using substance flow analysis

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Abstract

The European Union (EU) Water Framework Directive (WFD) requires Member States to protect inland surface and groundwater bodies but does not directly stipulate how the associated environmental quality standards should be achieved. This paper develops and assesses the performance of a series of urban emission control strategies (ECS) with an emphasis on the scientific and technological benefits which can be achieved. Data from the literature, in combination with expert judgement, have been used to develop two different semi-hypothetical case cities (SHCC), which represent virtual platforms for the evaluation of ECS using substance flow analysis (SFA). The results indicate that the full implementation of existing EU legislation is capable of reducing the total emissions of cadmium (Cd) and mercury (Hg) by between 11% and 20%. The ability to apply voluntary reduction practices is shown to be particularly effective for Cd with the potential to further lower the overall emissions by between 16% and 27%. The most efficient protection of the receiving surface water environment is strongly influenced by the city characteristics with the introduction of stormwater treatment practices being particularly effective for one city (59% reduction of Hg; 39% reduction of Cd) and the other city being most influenced by the presence of efficient advanced wastewater treatment processes (63% reduction of Hg; 43% reduction of Cd). These reductions in receiving water loads are necessarily accompanied by either increases in stormwater sediment loadings (2.6 to 14.9 kg/year or 0.6 to 2.4 kg/year for Hg) or wastewater sludge loadings (45.8 to 57.2 kg/year or 42.0 to 57.4 kg/year for Cd).

Keywords: Priority pollutants; semi-hypothetical case cities; substance flow analysis; cadmium; mercury.

1. Introduction

In Europe, point source discharges have been targeted through the implementation of several European Union (EU) Directives (e.g. EU Integrated Pollution Prevention and Control (IPPC) Directive (2008) and the EU Urban Waste-Water Directive (UWWD) (1991)) and the need to protect aquatic ecosystems through the mitigation of both point and diffuse urban pollution sources is being addressed through the implementation of EU Water Framework Directive (WFD) (2000). Measures set out in the WFD include reducing emissions of specified priority substances (PS) and the phase-out of discharges of priority hazardous substances (PHS) to achieve a good ecological and chemical status with environmental quality standards (EQS) having been established to support the attainment

of this goal (EU Environmental Quality Standards Directive, 2008). However, whilst point source pollutants such as industrial facilities and municipal wastewater treatment plants (WWTP) can be relatively easy to identify (and appropriate emission control measures developed), diffuse sources can be more difficult to locate and control, particularly in urban areas which are spatially and temporally dynamic.

The work described in this paper identifies and compares alternative strategies which can be used to mitigate the emissions of PS/PHS to receiving water bodies. Due to the unavailability of complete data sets for individual cities, an innovative approach is adopted involving the development and use of semi-hypothetical case cities (SHCC). These are virtual cities in which the parameters necessary to inform a comparison of alternative emission control strategies (ECS) are defined using a combination of real city data, literature data and, where neither of these is available, expert judgement to reduce the overall level of uncertainty in the final assessment.

Environmental control strategies can provide an important input into the establishment of programmes of measures (POMs) (Baarner, 2011) in order to improve water status at river basin catchment level. The POMs need to address the pressures arising from both urban and non-urban sources and their planning and implementation needs to involve a diverse range of stakeholders from different levels of government, industry, business and community. Ellis and Mitchell (2006) have specifically addressed the problems associated with tackling diffuse urban water pollution sources and Crabtree et al (2009) have assessed the benefits of pollutant reductions in both end-of-pipe discharges and diffuse sources. The substance flow analysis (SFA) approach, discussed in this paper, provides an alternative methodology for determining the impacts of different strategies to reduce pollutant loads derived from point and diffuse sources as well as recognising the importance of legislation and voluntary controls.

SFA supports an overall management strategy by identifying and quantifying inflows, stocks and outflows (e.g. exports and environmental emissions) of a single substance or group of substances within a defined system (van der Voet, 2002). It requires the definition of clear physical boundaries and an identified time frame which is normally one year. Burstrom (1999) and Linqvist and von Malmborg (2004) have pointed out the benefits of the SFA approach to environmental management at the municipal level based on the provision of both quantitative and qualitative information. Important compartments at the urban scale include air, soil, impermeable surfaces, groundwater, surface waters, sewage sludge and industrial waste. These are all considered in this paper but the focus is on the urban water system and the roles of wastewater and stormwater in transferring pollutants to the receiving water environment. Previous applications of SFA with respect to wastewater (Eriksson et al, 2008) and stormwater (Ahlman and Svensson, 2002; Bjorklund, 2010) have been reported for selected organic and inorganic pollutants.

This paper presents the SFA results for two PHS (cadmium (Cd) and mercury (Hg)) within two SHCC possessing different characteristics and uses these to inform a comparative evaluation of six alternative ECS which are designed to take account of different legislative, technical and voluntary approaches for minimising the releases of PS/PHS.

2. Methodology and implications of introducing different emission control strategies

2.1 Characteristics of hypothetical case study cities

The characteristics of the SHCC, required to inform the evaluative comparison of alternative ECS through the use of SFA, have been identified and defined through the collation of data from a range of sources. A total of 87 EU water-related research projects were reviewed to identify key urban descriptors (Eriksson et al., 2009), which were then quantitatively benchmarked based on a review of the literature, including the Eurostat statistical data set (2012), the Urban Audit (2008), and the use of expert judgement derived through discussions held within the EU FP6 ScorePP project. Using this approach the data for two SHCC, identified as an Eastern European Inland (EI) city and a Northern European Coastal (NC) city, have been collected. Key characteristics of EI are that it has a growing economy with many heavy industries, is experiencing substantial population growth and is situated by a large river. NC has a consumer-oriented industry, an environmentally-aware public sector and is located on the coast at the mouth of a medium-sized river (Eriksson et al., 2012). Data on the pathways, release factors and loads of Cd and Hg generated by identified sources have been derived from a previously established source classification database (Holten Lützhøft et al., 2012). Pollutant releases from specific activities and processes have been appropriately scaled-up or –down (e.g. according to population size, tonnes of municipal waste produced etc.) to yield SHCC specific loads (Eriksson et al., 2011).

2.2 Pollutant processes

The pollutant inputs to the urban water system originate from the process outflows producing PS/PHS during the chosen time period and from the products and commodities containing the PS/PHS. The pollutant pathways involving different components of the urban water environment are shown in Figure 1. The pollutant loads deposited on urban surfaces are considered to be washed off during storm events ending up in either the combined sewer system or the storm sewer system with the latter being treated in stormwater best management practice systems (BMP) or discharged directly to a receiving water environment. Within a stormwater BMP, the PS/PHS can be biotically or abiotically degraded or accumulated in sediments prior to discharge. Pollutants entering the combined sewer may be treated at the WWTP (involving processes such as biodegradation, volatilisation, and sludge accumulation) or discharged to the river through combined sewer overflows during times of high flow. The involvement of the different pathways will be dependent on both the characteristics of the SHCC and the applied ECS but within the urban water system the following represent important receiving ‘compartments’:

- Air (volatilization)
- Sludge (e.g. at the WWTP)
- Sediment (e.g. within stormwater BMP)
- Soil (direct deposition or accumulation during infiltration)
- Groundwater (following infiltration)
- Discharge to receiving waters.

2.3 Emission control strategies (ECS)

The potentials for the removal of Cd and Hg from urban water systems in the two SHCC are investigated through the application of six different ECS. ECS1 represents the current or ‘business-as-usual’ scenario for which the relevant contributions to the different water compartments and the resulting percentage metal removals are shown in Table 1. For EI, typically 50% of the rainfall-

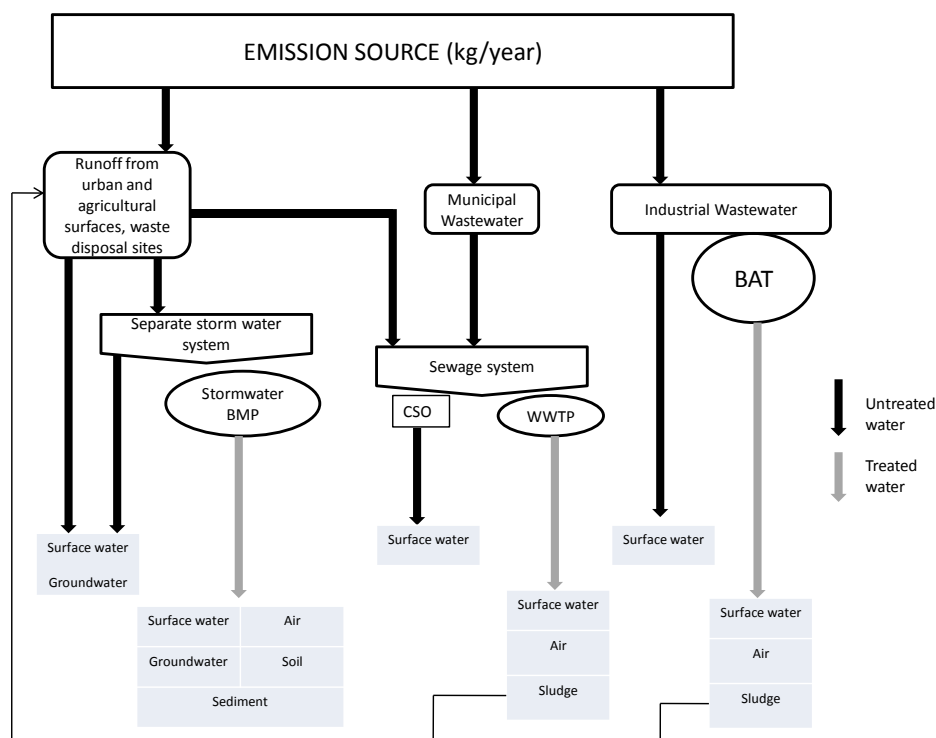


Figure 1. The pollutant pathways in the urban water environment considered in the SFA process.

runoff enters the stormwater system but this reduces to 10% for NC and in both cases it is estimated that only 25% of the stormwater receives treatment through the use of detention/retention ponds. Hossain et. al. (2005) have reported total Cd removal efficiencies for a highway stormwater pond can vary from 0 to 100% with a mean of 55%. A consideration of the theoretical implications for Cd removal in pond systems suggests a lower efficiency (Revitt et. al., 2008) and 30% has been used in this application (Table 1). The predicted removal efficiencies for the same treatment systems for Hg are higher at 70-90% (Revitt et. al., 2008) with measured values for an urban stormwater pond being quoted as 56% (Semadeni-Davies, 2006). A compromise value of 70% has been used resulting in the Hg removal potentials shown in Table 1. Under ECS1, 18% of the stormwater entering the combined sewer system, is estimated to be discharged direct to surface waters for EI compared to 10% for NC.

The percentages of municipal wastewaters which are assumed to be treated to secondary standard are shown in Table 1 with this level of treatment being considered to lead to the incorporation of an average of 60% Cd (Buzier et.al., 2006; Karvelas et.al., 2003) and 80% Hg (Balogh and Liang, 1995; Mugan, 1996) into WWTP sludge. Under ECS1, it is recognised that only the heavy polluting industries receive treatment to the level demanded by EU IPPC Directive (2008) under the Best Available Technology (BAT) philosophy leading to the low percentages of industrial wastewaters which receive effective treatment (Table 1). From the range of BAT techniques available for the treatment of both Cd and Hg, a combination of precipitation and settling is considered appropriate to remove Cd in both cities resulting in a 70% reduction in the discharges to water (EC Reference Document on Ceramic Manufacturing Industry, 2007). In the case of Hg, settling/clarification are the main removal processes employed in EI (70% reduction) whereas more sophisticated techniques involving nanofiltration/adsorption are utilised in NC producing 90% removal of Hg from industrial wastewaters. The resulting small overall reductions in industrial metal discharges are shown in Table 1.

Table 1. Parameters used to identify the behaviours of Cd and Hg in urban water systems in EI and NC for the baseline Emission Control Strategy (ECS1).

	EI		NC	
	Cd	Hg	Cd	Hg
Percentage of stormwater discharged to separate sewer	50		10	
Percentage of stormwater which is treated	25		25	
Percentage of metal removed from treated stormwater	7.5	17.5	7.5	17.5
Percentage of stormwater discharged to combined sewer	50		90	
Percentage of wastewater treated to secondary standard	78		95	
Percentage of metal removed at WWTP	46.8	62.4	57	76
Percentage of wastewater discharged as combined sewer overflows	18		10	
Percentage of industrial wastewater receiving effective treatment	3.0		2.3	
Percentage of metal removed by industrial wastewater treatment	2.1	2.1	1.6	2.1

ECS2 relates to the situation which would be achieved following the full implementation of appropriate existing EU legislation. Full details of the relevant EU Directives are provided in Table 2 together with the predicted impacts on the emissions of Cd and Hg which are based on whether a complete pollutant ban, a use restriction or the achievement of an emission limit is required and taking into account the usage or process applications in the different cities. Also in ECS2, it is assumed that all municipal wastewaters are treated to at least secondary standard in line with the requirements of the EU UWWTD (1991). This has the effect of increasing the Cd and Hg loads removed to sewage sludge at the WWTPs of both cities to 60% and 80% of the inputs, respectively. The metal removal efficiencies for Cd and Hg achieved from industrial wastewaters by BAT are extended to moderately and heavily polluting industries with the consequence that 11.6% and 8.7% of industrial discharges will be efficiently treated in EI and NC, respectively. The resulting metal reductions are 8.5% for Hg and Cd in EI, 7.8% for Hg NC and 6.1% for Cd in NC.

ECS 3-6 incorporate the measures already discussed and implemented under ECS2 with additional control actions according to the main sources of the pollutants. ECS3 is characterised by the inclusion of a range of voluntary mitigation options at the household, commercial and/or industrial levels as identified in Table 3 together with the predicted additional reductions. In many cases the reductions are difficult to identify precisely because, being voluntary, they will be influenced by the success of stakeholder-driven initiatives such as eco-labelling practices, information/education campaigns and financial incentives to promote reuse and recycling. Many of the available options relate to source control measures involving substitution of the item containing the pollutant (e.g. artist paints containing Cd) or the management of processes that may result in release (e.g. Hg deriving from dental amalgam). Mitigation of releases from existing stocks can be achieved by careful disposal of waste leading, for example, to lower environmental releases as can be seen in Table 3 with, 20-25% reductions in Cd emissions being achievable through the effective implementation of battery recycling at household and commercial levels.

Table 2. Predicted percentage reductions in the respective contributions of Cd and Hg in EI and NC as a consequence of implementation of existing EU Directives (ECS2).

Relevant EU Directive	Predicted percentage reductions in emissions to the water environment.			
	Cd		Hg	
	EI	NC	EI	NC
EU Fertiliser Directive. 1976 (Directive 76/116/EEC). http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:31976L0116:EN:HTML ; places restrictions on selected metals in fertilisers	25	25		
EU Sewage Sludge Directive. 1986. (Directive 86/278/EEC). http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:31986L0278:EN:HTML ; restricts metals in sewage sludge used for agriculture	25	25	20	25
EU Batteries and Accumulators Directive. 1991. (Directive 91/157/EEC). http://eur-lex.europa.eu/Result.do?T1=V1&T2=1991&T3=157&RechType=RECH_naturel&Submit=Search ; regulates the management of batteries containing >0.25% Cd	25	25		
EU Dangerous Substances Directive. 1991. (Directive 91/338/EEC). http://eur-lex.europa.eu/Result.do?T1=V1&T2=1991&T3=338&RechType=RECH_naturel&Submit=Search ; prohibits Cd use in paints and Hg in textile preservatives under restrictions on the marketing and use of certain dangerous substances and preparations.	50	50	20	20
EU Foodstuff Contamination Regulation. 2001. (Regulation (EC) No 466/2001). http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:32001R0466:EN:HTML ; sets maximum levels for Cd in foodstuffs	25	25		
EU Waste Electrical and Electronic Equipment (WEEE) Directive. 2002. (Directive 2002/96/EC). http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:32002L0096:EN:HTML ; sets limits on Hg release from waste electrical and electronic equipment			20	25
EU Waste Landfill Directive. 1999. (Directive 1999/31/EC) on the landfill of waste. http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:31999L0031:EN:HTML sets limits regarding Hg discharge from landfill			15	20
EU Hazardous Waste Incineration Directive. 1994. (Directive 94/67/EC); http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:31994L0067:EN:HTML ; requires incineration of Hg containing waste with limited harm to the environment			15	20
EU Mercury in Measuring Devices Directive. 2007. (Directive 2007/51/EC). http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2007:257:0013:01:EN:HTML ; restricts the use of Hg in measuring devices.			25	25
EU Plant Protection Product Directive. 1979. (Directive 79/117/EEC). http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:31979L0117:EN:HTML ; prohibits the use of Hg in plant protection products			25	25
EU Large Combustion Plant Directive. 2001. (Directive 2001/80/EC). http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:32001L0080:EN:HTML ; restricts emissions from large combustion plants			20	20

ECS4 applies specifically to the industrial processes resulting in discharges of Cd and/or Hg in each SHCC and considers the impact of the widespread application of on-site treatment using BAT. All industrial facilities are included regardless of size and polluting potential and the envisaged removal efficiencies are increased from 70 to 80% for Cd in both cities and for Hg in EI. The corresponding value for Hg in NC is maintained at 90% as was applied to the smaller number of industries included in ECS1 and ECS2. The identified increases in removal efficiencies are due to the feasibility of extending the industrial wastewater treatment techniques beyond generic BAT and are consistent with the range of removal efficiencies in excess of 90% which have been reported for both Cd (Rao et. al., 2010) and Hg (Barron-Zambrano et. al., 2004).

ECS5 represents improvements to the management of stormwater and the mitigation of combined sewer overflows through the use of stormwater BMPs. These treatment systems are considered to be applied to 80% and 90% of the discharged stormwater in EI and NC, respectively. In addition, by installing BMPs such as infiltration basins, constructed wetlands and porous paving, which have been shown to be efficient at removing metals (Revitt et al, 2008), the removal efficiencies can be

increased to 70% for Cd (Visesmanee et. al., 2008) and 90% for Hg (Nelson et. al., 2006). Consequently, the overall metal removal levels which can be achieved are 56% and 63% for Cd in EI and NC and 72% and 81% for Hg in EI and NC.

Table 3. Predicted percentage reductions in the respective contributions of Cd and Hg in EI and NC as a consequence of applying voluntary reductions (ECS3).

Details of identified voluntary reduction practices	Percentage reduction contributions additional to ECS2			
	Cd		Hg	
	EI	NC	EI	NC
Recycling at the household level	5	10	10	15
Specific household/commercial reductions				
• Domestic and artist paints	20	25		
• Battery recycling	20	25		
Collection and treatment of waste	25	40	10	10
Waste incineration	10	20		
Specific industrial reductions				
• Dental Hg traps			40	50

The use of advanced wastewater treatment technologies beyond the current legislative requirements (ECS2) is envisaged under ECS6. In addition to the cities being fully served by wastewater treatment to primary and secondary level some of the treatment plants in this scenario are equipped with appropriate tertiary treatment or extended sludge aging to remove the relevant priority substances. For EI it is assumed that 50% of the municipal sewage is treated to tertiary level and that this increases to 75% for NC. Based on improved overall Cd removal efficiencies to 90% when using tertiary treatment (Wu et. al., 2005), the Cd reductions achieved at the WWTP would increase to 75% and 87.5% for EI and NC, respectively. The corresponding values for Hg, based on 95% removal by tertiary treatment would be 87.5% and 91.2%.

To be consistent with the stated objectives, the emphasis behind the development of the different ECS has concentrated on scientific and technical considerations. However, reference has also been made to the relevance of political aspects (e.g. ECS2) and to the importance of organisational/operational capabilities (e.g. ECS3) in contributing to the reductions in Cd and Hg emissions. Financial aspects (including capital, operational and maintenance costs) will also influence the feasibility of introducing a particular ECS but are difficult to assess partly because of the different ways in which the economic costs are presented. Viavattene et al (2010) have reviewed the limited economic data which is available in relation to the introduction of technical options (ECS4-6) The cost implications need to be balanced against the identified benefits e.g. the social awareness improvements associated with the installation of BMPs to reduce the impacts of stormwater (ECS5) and the environmental gains deriving from the instigation of voluntary controls.

It is clear that the definitive selection of a control strategy requires the active involvement of a number of different stakeholders whose views reflect a diverse range of priorities and concerns. To define the most appropriate ECS as part of a collective policy-making and implementation process becomes complex and intuitive reasoning and expert judgement may not be sufficient to achieve a decision that satisfies all stakeholders (Brunner and Starkl, 2004). One approach for ensuring transparency and accountability in reaching a decision is the use of multi-criteria analysis (MCA) in which the performances of several options are assessed against a range of established criteria. Indicators can then be developed to provide a measure of the extent to which the criteria are met by the selected options ([Ellis et al., 2008](#); Makropoulos et al., 2008) through the completion of a bench-

marking exercise which enables the decision-maker to compare alternatives in a robust, transparent and accountable way. Table 4 identifies a range of criteria and supporting indicators which could act as a framework to support a more holistic integrated evaluation of ECS with regard to their impact on substance flow. This paper concentrates on technical feasibility, efficiency of proposed procedure and environmental impact with less emphasis on economic considerations and social impact.

Table 4. Criteria and associated indicators for assessing the technical and socio-economic benefits of a proposed emission control scenario (ECS)

Criteria	Indicators
Technical feasibility	Extent to which appropriate technology exists
	Extent to which appropriate technology will exist by 2015
Efficiency of proposed procedure	Confidence with which EQS can be achieved by 2015 through reductions of priority substances
	Confidence with which cessation of emissions can be achieved by 2025
Economic considerations	Level of capital costs involved (including adaptation)
	Level of operating and maintenance costs
	Level of additional cost benefits
Environmental impact	Level of impact on ecological status of receiving water body
Social impact	Changes to community/amenity benefits
	Extent to which labour market is affected
	Impact of possible non-product availability

3. Results and Discussion

3.1 Overall emissions of Cd and Hg

Mercury is emitted from a range of point and diffuse sources, of which 37 different sources have been identified in EI and NC including combustion (e.g. transport, energy production), industrial processes by-products (e.g. metals and paper), dental amalgam and various household goods (Eriksson et al., 2011¹⁰). Cadmium is ubiquitous in the urban environment and 55 sources have been identified in EI and NC, ranging from transportation and electricity generation to the life-cycle of Cd-containing products such as stabilisers, alloys and electrical components (van de Voet et. al., 1994).

Table 5 gives an overview of the predicted total emissions of Hg and Cd to the environment in both cities following the application of SFA to each ECS. Under the current existing situation (ECS1), the total emissions of Hg to the environment are higher in NC than EI. In contrast, the total emissions of Cd are greater in EI in comparison to NC, reflecting differences in city-specific characteristics such as the numbers and types of local industries, population size and waste management practices. The full implementation of existing European legislation (ECS2) has the potential to reduce total emissions to the urban environment by 11% (Hg in EI), 20% (Hg in NC) and 18% (Cd in both cities) (Table 5). The impact of the introduction of voluntary mitigation options (ECS3), such as the installation of dental amalgam traps and the availability of efficient battery collection programmes further reduces the total emissions of Cd and Hg in both cities. However, this scenario is considerably more beneficial for Cd as the potential emission reduction measures are directed at more numerous widely practised sources. The reductions of Cd emissions due to the adoption of voluntary activities are predicted to be considerably greater for NC (27%) compared to EI (16%) with one reason being the more socially aware attitudes of the population in the former city. The installation of state-of-the-art water treatment facilities for industrial wastewater, stormwater and municipal wastewater (ECS4-6) does not have the potential to reduce the overall emissions of Cd

and Hg for either city above the level achieved by ECS2 (Table 5) but they do exert an important influence on the location of the final environmental compartment.

Table 5 Total emissions of Hg and Cd to the environment in EI and NC (kg/year)

	Total emissions of Hg (kg/year)		Total emissions of Cd (kg/year)	
	EI	NC	EI	NC
ECS1	135	172	613	480
ECS2	120	138	503	392
ECS3	110	130	423	285
ECS4, 5 and 6	120	138	503	392

3.2 Emissions to individual environmental compartments

To examine the overall trends in more detail, the SFA approach has been used to assess the Cd and Hg loads emitted to the individual environmental compartments identified in Figure 1. Emissions to surface waters may occur directly from the source or in the discharges arising from untreated urban runoff and combined sewer overflows and to a lesser extent in the effluents from treated industrial and municipal wastewaters. Direct emissions of Cd to water occur following the application of fertilisers (Cupit et. al., 2002; Thevenot et. al., 2007) during market gardening activities and the landfilling of wastes collected from, for example, the production of Ni/Cd batteries. Comparable Hg emissions occur due to the erosion of car tyres and road surfaces (Bergback et. al., 2001) as well as from coal combustion installations (Sundseth et. al., 2012). Landfill activities (Al-Muzaini, 2009) as well as the spreading of sewage sludge to agricultural land (McBride et. al., 1999) have been identified as sources of both metals to groundwaters. The emphasis on sludge disposal to land (50% of the total in NC; 60% of the total in EI) also contributes to the build up of both metals in the soil environment (Thevenot et. al, 2007; Carbonell, 2009). The sludge environmental compartment is added to as a result of both industrial and wastewater treatment processes with dental practices making an important Hg contribution (Bergback et. al., 2001). BMPs represent an important treatment facility for stormwater with the sediment compartment acting as a sink for Cd and Hg. Municipal waste incineration is an important contributor to the atmospheric compartment in both cities. Additionally, Cd is emitted to air by Cd processing plants and the manufacture of Ni/Cd batteries (Pacyna, 1984) with airborne Hg arising from coal combustion (Pirrone et. al., 2010), crematorium incinerators (Bergback et. al., 2001; Takaoka et. al., 2010) and landfill emissions (Holten Lützhøft et al., 2012).

3.2.1 Analysis of Hg emissions

The patterns of Hg emissions to the different identified environmental compartments are shown in Figures 2 and 3 for EI and NC, respectively. Major differences can be observed under the existing scenario (ECS1) with emissions to surface water dominating in EI (Figure 2) followed by emissions to WWTP sludge and soil which are at lower but comparable levels. This contrasts sharply with the pattern of Hg emissions in NC, where emissions to air are highest, followed by emissions to WWTP sludge and then emissions to surface water (Figure 3). Whilst the larger population in EI would be expected to produce greater Hg emissions to air from cremations, it appears that a more important influencing factor for this environmental compartment is the higher amount of land-filled waste produced by the increased consumer related activities in NC compared to EI (Eriksson et al., 2012). Hg emissions from landfill are associated with the production and subsequent release of methyl mercury in landfill gases (Lindberg et al., 2005).

Within EI, the Hg emission patterns show clear variations due to the progressive applications of ECS 2-6. The largest decrease is observed in the release of Hg to receiving waters due to the application of ECS5 (ECS2 combined with the widespread use of stormwater BMPs (Figure 2)). This reduction in Hg emissions to water (59%) is related to the fact that diffuse sources such as heating and traffic make important contributions of Hg to stormwater runoff prior to removal in ECS5. In NC, the Hg emissions to air and water show the most significant variations, with decreases in the former (25%) being solely influenced by the implementation of the relevant legislation (ECS2). The largest reductions in Hg emissions to surface waters (63%) are associated with the improvements in WWTP efficiencies (ECS6) (Figure 3).

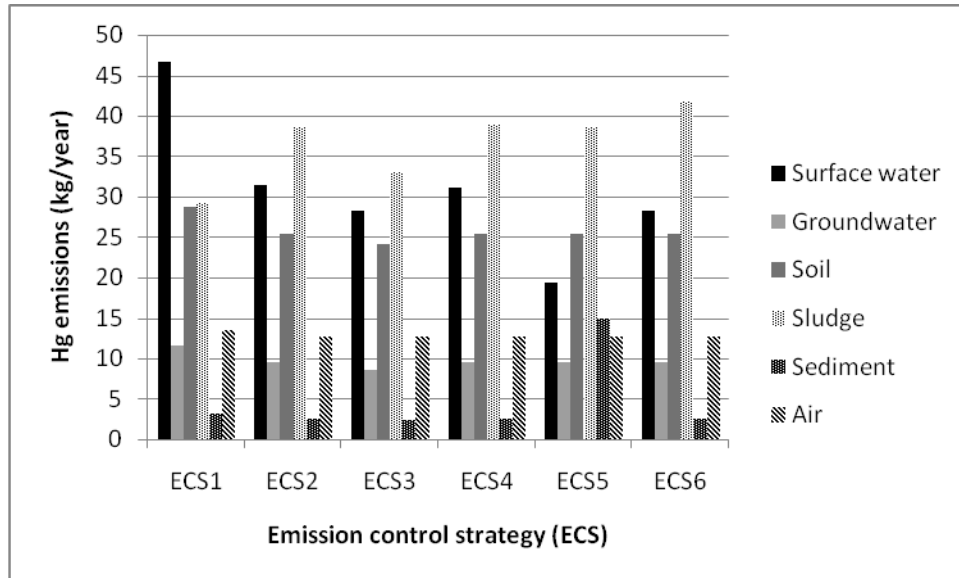


Figure 2. The relative distribution of Hg between the different environmental compartments in EI (kg/year)

The Hg emissions to all environmental compartments generally decrease in both cities as a consequence of the application of ECS2. However, an exception is the emission to the wastewater treatment sludge compartment in EI where a 32% increase is predicted reflecting the impact of, for example, increasing the number of properties connected to the WWTP. Other increases occur depending on which water treatment practices exert important influencing roles by reducing the Hg emissions to receiving waters. This is illustrated by the role of ECS5 in EI, which results in a quadrupling of emissions to the stormwater BMP sediment. In NC, the impact of ECS6 produces a 8% increase in Hg loading to WWTP sludge. The translocation of Hg between different compartments is consistent with the fact that as a metal, Hg cannot be degraded (although its compounds can be biologically and chemically transformed).

3.2.2 Analysis of Cd emissions

The SFA results for Cd for ECS1 show elevated emissions to the majority of individual environmental compartments compared to Hg but the overall trends in both cities (Figures 4 and 5) are similar. The impacts appear to be greater in EI due to the higher Cd emissions in this city. Emissions to groundwater dominate under all ECS followed by emissions to soil due to activities

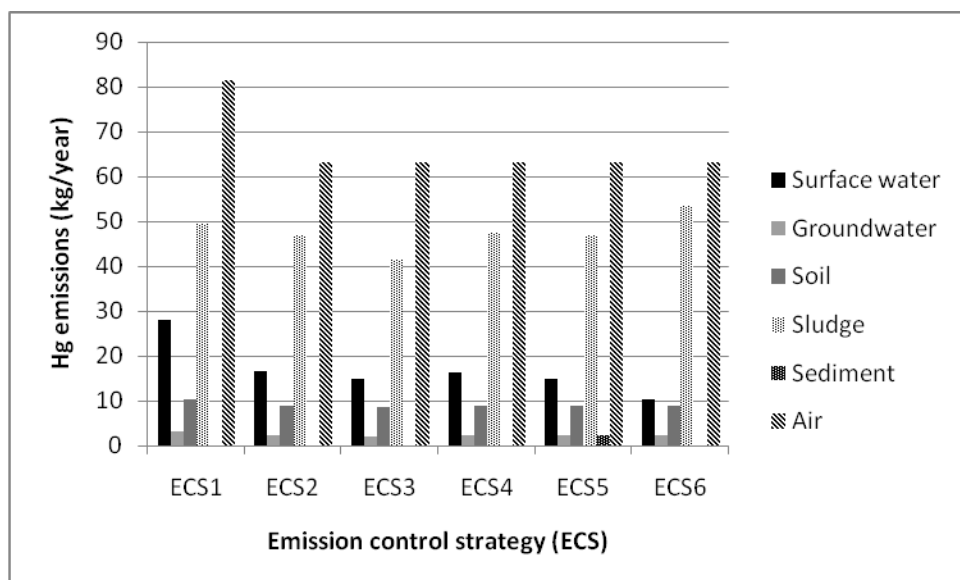


Figure 3. The relative distribution of Hg between the different environmental compartments in NC (kg/year)

such as land filling of municipal waste, waste incineration and the use of fertilisers in both cities. The higher Cd loads entering groundwater are consistent with the established mobility of this metal in the soil environment (Dusek et. al., 2010). The lowest emissions of Cd in both cities are consistently to sludge, sediments and air. This is particularly noticeable in sediments except for the widespread implementation of BMPs which leads to elevated levels in treatment system sediments. The lower atmospheric presence of Cd compared with Hg is consistent with its lower volatility.

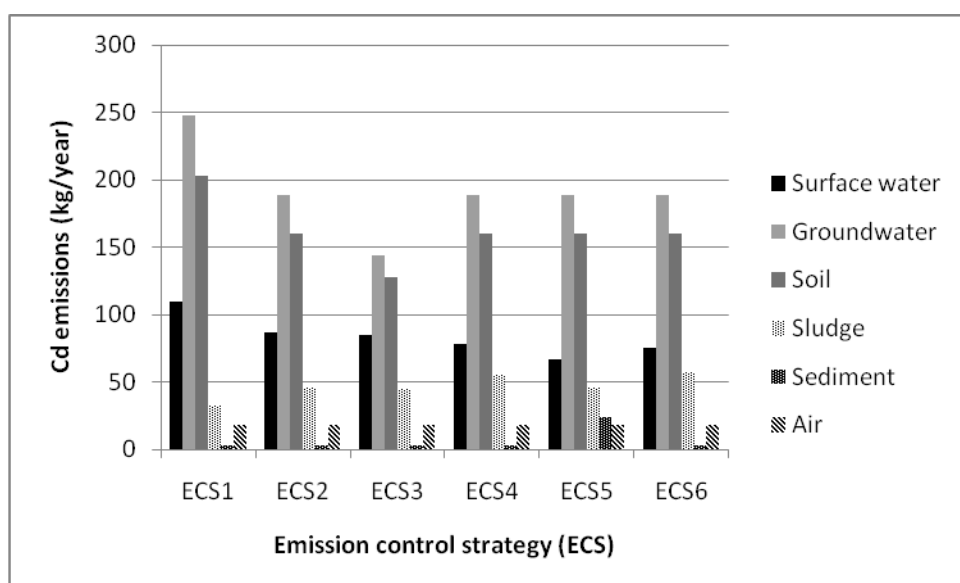


Figure 4. The relative distribution of Cd between the different environmental compartments in EI (kg/year)

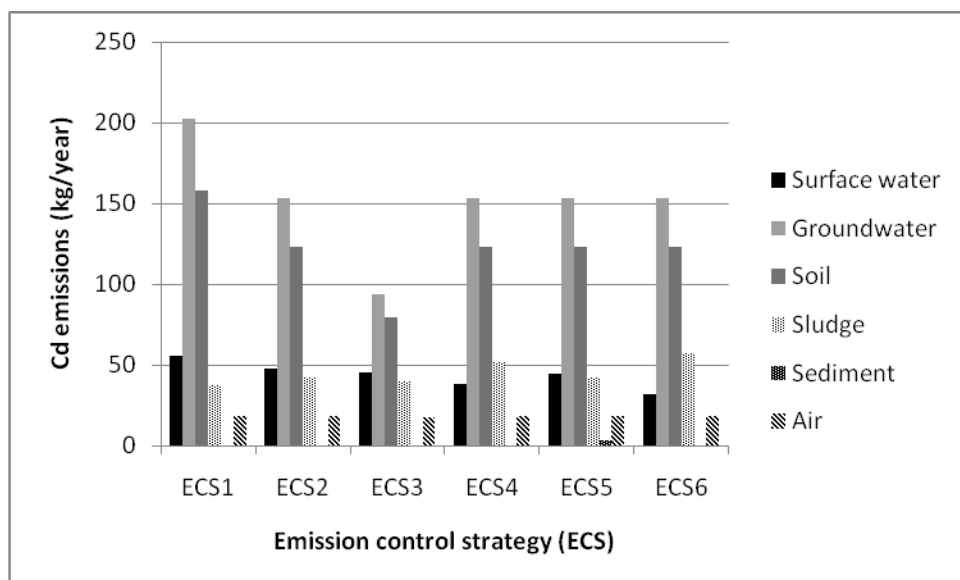


Figure 5. The relative distribution of Cd between the different environmental compartments in NC (kg/year)

The applications of ECS2 and ECS3 significantly reduce the Cd loads to soil (37% to 49%) and groundwater (42% to 54%) but have a lesser impact on emissions to water (19 to 22%). However, further reductions in Cd discharges to receiving waters occur with the introduction of extended and improved water treatment processes (ECS4-6) but, as for Hg, it is the widespread use of stormwater treatment which appears to be most effective in EI achieving a 39% reduction. The corresponding Cd reduction in NC is only 20% but in this city better protection of the receiving water environment is offered by improved treatment of industrial wastewaters (32% reduction) and advanced municipal wastewater treatment (43% reduction). As was observed for Hg emissions, the reductions in Cd discharges to surface waters are accompanied by increases in the Cd loads found in BMP sediments (640%) and WWTP sludges (54%). The increase in sludge loads may be associated with a combination of factors including the increased connection of homes to a WWTP under ECS2 in EI and the use of Cd in a wider range of products in both cities in comparison to Hg. The treatment technologies identified in ECS4-6 result in the translocation of the metal rather than a reduction to the environment as a whole.

4. Conclusions

In the EU, the implementation of the WFD presents a major challenge which is being addressed through the development and implementation of POMs to achieve the stringent objectives. In the absence of complete data sets on catchment characteristics and the efficiencies of treatment technologies for the removal of PS/PHS, the implementation of alternative ECS within SHCC platforms is proposed as an approach to support practitioners to achieve the legislative requirements by using a combination of available data and (in its absence) expert judgement. The presented results identify the potential of using SFA within a SHCC environment to distinguish, at a sufficiently sensitive level, between the applications of alternative ECS on a 'city-by-city' and 'pollutant-by-pollutant' basis. For the chosen pollutants of Cd and Hg, the selected cities are shown to demonstrate preferential benefits from the implementation of different water treatment options with the extent of these being dependent on the pollutant. The essentially non-destructible characteristics of Cd and Hg results in their movement between compartments, with reductions in the emissions to one

compartment corresponding to increased emissions to another. This represents an important policy consideration which needs to be taken into account, and be informed by scientific opinion, when assessing the most appropriate implementation option. Reductions in total emissions to the urban environment can only be achieved through targeting the use of Cd and Hg ‘at source’ through for example, the application of legislation (ECS2) and/or voluntary measures (ECS3) to limit their presence in the supply chain and resulting products. Currently, the application of the mass-balance approach includes limited consideration of socio-economic factors. The fuller inclusion of these aspects is important in terms of informing the development of POMs which are economically feasible and socially acceptable to industry, commercial operators and wider society.

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References

- Ahlman, S. and Svensson, G., 2002. Substance flow analysis of the stormwater system in Vasastaden, Goteborg. *Global Solutions for Urban Drainage. Proceedings of 9th International Conference on Urban Drainage*. doi:10.1061/40644(2002)100
- Al-Muzaini, S. M. 2009. A comparative study of the characterization of landfill leachate at the dumping sites in Kuwait. *J Food Agric Environ*, 7, 679-683.
- Baarnier, L., 2011. The programme of measures of the Water Framework Directive – more than just a formal compliance tool. *J Eur Environ Plan Law*, 8, 82-100.
- Balogh, S. J. and Liang, L. 1995. Mercury pathways in municipal wastewater treatment plants. *Water Air Soil Poll*, 80, 1181-1190.
- Barron-Zambrano, J., Laborie, S., Viers, Ph., Rakib, M. and Durand, G. 2004. Mercury removal and recovery from aqueous solutions by coupled complexation-ultrafiltration and electrolysis. *J Membrane Sci*, 229, 179-196.
- Bergback, B., Johansson, K. and Mohlander, U. 2001. Urban metal flows—A case study of Stockholm. *Review and Conclusions. Water Air Soil Poll Focus*, 1, 3-24.
- Bjorklund, K., 2010. Substance flow analyses of phthalates and nonylphenols in stormwater. *Water Sci Technol*, 62, 1154-1160.
- Brunner, N. and Starkl, M. 2004. Decision aid systems for evaluating sustainability: a critical survey. *Environ. Impact Assess. Rev.*, 24(4), 441-469.
- Burstrom, F., 1999. Materials accounting and environmental management in municipalities. *J Environ Assess Pol Manag*, 1, 297-327.

- Buzier, R., Tusseau-Vuillemin, M. H., Meriadec, C. M. D., Rousselot, O. and Mouchel, J. M. 2006. Trace metal speciation and fluxes within a major French wastewater treatment plant: Impact of the successive treatments stages. *Chemosphere*, 65, 2419-2426.
- Carbonell, G., Pro, J., Gomez, N., Babin, M. M., Fernandez, C., Alonso, E. and Tarazona, J. V. 2009. Sewage sludge applied to agricultural soil: Ecotoxicological effects on representative soil organisms. *Ecotox Environ Safe*, 72, 1309-1319.
- Crabtree, B., Kelly, S., Green, H., Squibbs, G. and Mitchell, G., 2009. Water Framework Directive catchment planning: a case study apportioning loads and assessing environmental benefits of programme of measures. *Water Sci Technol*, 59(3), 407-416.
- Cupit, M, Larsson, O., de Meeu's, C, Eduljee, G. H. and Hutton, M. 2002. Assessment and management of risks arising from exposure to cadmium in fertilisers—II. *Sci Tot Environ*, 291, 189–206
- Dusek, J., Vogel, T., Lichner, L. and Cipakova, A. 2010. Short-term transport of cadmium during a heavy-rain event simulated by a dual-continuum approach. *J Plant Nutr Soil Sci*, 173, 536-547.
- EC Reference Document on Ceramic Manufacturing Industry. 2007. Best Available Techniques in the Ceramics Manufacturing Industry. http://eippcb.jrc.es/reference/BREF/cer_bref_0807.pdf (last accessed 1 March 2012).
- Ellis, J.B., Deutsch, J-C, Legret, M., Martin, C., Revitt, D.M., Scholes, L., Seiker, H. & Zimmerman, U., 2006. The DayWater decision support approach to the selection of sustainable drainage systems. *Water Practice and Technology* 1 (1),8.
- Ellis, J. B. And Mitchell, G., 2006. Urban diffuse pollution: key data information approaches for the Water Framework Directive. *Water Environ J*, 20(1), 19-26.
- Eriksson, E., Anderson, H. R. and Ledin, A., 2008. Substance flow analysis of parabens in Denmark complemented with a survey of presence and frequency in various commodities. *J Hazard Mater*, 156, 240-259.
- Eriksson, E., Donner, E., Raggatt, L., Pettersson, M., Wickman, T. and Mikkelsen, P. S. 2009. Strategies for controlling emissions of priority pollutants from case city archetypes. In *Proceedings of WEFTEC 09, 82nd Annual Water Environment Federation Technical Exhibition and Conference*, Orlando, Florida, USA, 14 October 2009.
- Eriksson, E., Revitt, D. M., Ledin, A., Lundy, L., Holten Lützhøft, H. C., Wickman, T. and Mikkelsen, P. S. 2011. Water management in cities of the future using emission control strategies for priority hazardous substances. *Water Sci Technol*, 64, 2109-2118.
- Eriksson, E., Revitt, M., Holten-Lützhøft, H.C., Viavattene, C., Scholes, L. and Mikkelsen, P.S. 2012. Emission control strategies for short-chain chloroparaffins in two semi-hypothetical case cities. In: Rauch, S., and Morrison, G.M. (Editors), *Urban environment: Proceedings of the 10th Urban Environment Symposium: Urban Futures for a Sustainable World*, Alliance for Global Sustainability Bookseries, Springer, Vol. 19. pp 213-223. (ISBN 978-94-007-2539-3).

EU Environmental Quality Standards (EQS) Directive. 2008. Directive 2008/105/EC of the European Parliament and of the Council on environmental quality standards in the field of water policy. <http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:32008L0105:EN:NOT> (last accessed 1 March 2012)

EU Integrated Pollution Prevention and Control Directive. 2008. Directive 2008/1/EC of the European Parliament and of the Council concerning integrated pollution prevention and control. <http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2008:024:0008:01:EN:HTML> (last accessed 1 March 2012)

EU Urban Waste-Water Directive. 1991. Directive 91/271/EEC concerning urban waste-water treatment. <http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:1991:135:0040:045:EN:HTML> (last accessed 1 March 2012).

EU Water Framework Directive (WFD). 2000. Directive 2000/60/EC of the European Parliament and of the Council establishing a framework for Community action in the field of water policy. <http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:32000L0060:EN:HTML> (last accessed 1 March 2012.)

Eurostat statistical databases. (2012). Available from <http://epp.eurostat.ec.europa.eu/portal/page/portal/statistics/themes>.

Holten Lutzhoft, H-C., Donner, E., Banovez, P., Wickman, T., Eriksson, E., Steen Mikkelsen, P.S., Ledin, A. 2012. A source classification framework supporting pollutant source mapping, pollutant release prediction, transport and load forecasting, and source control planning for urban environments. *Environ Sci Pollut Res*, 19, 1119-1130.

Hossain, M. A., Alam, M., Yonge, D. R. and Dutta, P. 2005. Efficiency and flow regime of a highway stormwater detention pond in Washington, USA. *Water Air Soil Poll*, 164, 79-89.

Karvelas, M., Katsoyiannis, A. and Samara, C. 2003. Occurrence and fate of heavy metals in the wastewater treatment process. *Chemosphere*, 53, 1201-1210.

Lindberg, S.E., Southworth, G., Prestbo, E.M., Wallschlager, D., Bogle, M.A. and Price, J. 2005. Gaseous methyl- and inorganic mercury in landfill gas from landfills in Florida, Minnesota, Delaware, and California. *Atmos Environ*, 39, 249-258.

Lindqvist, A. and von Malmberg, F., 2004. What can we learn from local substance flow analyses? The review of cadmium flows in Swedish municipalities. *J Clean Prod*, 12, 909-918.

Makropoulos, C.K., Natsis, K., Liu, S., Mittas, K. and Butler, D. 2008. Decision support for sustainable option selection in integrated urban water management. *Environ. Modell. Softw*, 23(12). 1448-1460.

McBride, M. B., Richards, B. K., Steenhuis, T. and Spiers, G. 1999. Long-term leaching of trace elements in a heavily sludge-amended silty clay loam soil. *Soil Sci*, 164, 613-623

Mugan, T. J. 1996. Quantification of total mercury discharges from publicly owned treatment works to Wisconsin surface waters. *Water Environ Res*, 69, 229-234.

Nelson, E. A., Specht, W. L. and Knox, A. S. 2006. Metal removal from water discharges by a constructed treatment wetland. *Eng Life Sci*, 6, 26-30.

Pacyna, J. F. 1984. Estimation of the atmospheric emissions of trace elements from anthropogenic sources in Europe. *Atmos Environ*, 18, 41-50.

Pirrone, N., Cinnirella, S., Feng, X., Finkelman, R. B., Friedli, H. R., Leaner, J., Mason, R., Mukherjee, A. B., Stracher, G. B., Streets, D. G., and Telmer, K. 2010. Global mercury emissions to the atmosphere from anthropogenic and natural sources. *Atmos Chem Phys*, 10, 5951–5964.

Rao, K., Mohapatra, M., Anand, S. and Venkateswarlu, P. 2010. Review on cadmium removal from aqueous solutions. *Int J Eng Sci Technol*, 7, 81-103.

Revitt, D. M., Scholes, L. and Ellis, J. B. 2008. A pollutant removal prediction tool for stormwater derived diffuse pollution. *Water Sci Technol*, 57, 1257-1264.

Semadeni-Davies, A. 2006. Winter performance of an urban stormwater pond in southern Sweden. *Hydrol Process*, 20, 165-182.

Sundseth, K., Pacyna, J. M., Pacyna, E. G. and Panasiuk, D. 2012. Substance flow analysis of mercury affecting water quality in the European Union. *Water Air Soil Poll*, 223, 429-442.

Takaoka, M., Oshita, K., Takeda, N and Morisawa, S. 2010. Mercury emission from crematories in Japan. *Atmos. Chem. Phys.*, 10, 3665-3671.

Thevenot, D. R., Moilleron, R, Lestel, L, Gromaire, M-C, Rocher, V, Cambier, P., Bonte, P. and Colin, J-L., Ponteves, C. and Meybeck, M. 2007. Critical budget of metal sources and pathways in the Seine River basin (1994–2003) for Cd, Cr, Cu, Hg, Ni, Pb and Zn. *Sci Tot Environ*, 375, 180-203.

Urban Audit. 2008. Measuring the quality of life in European cities.

http://epp.eurostat.ec.europa.eu/cache/ITY_OFFPUB/KS-SF-08-082/EN/KS-SF-08-082-EN.PDF
(last accessed 1 March 2012).

Van der Voet, E., van Egmond, L., Kleijn, R. and Huppes, G. 1994. Cadmium in the European Community: a policy-oriented analysis. *Waste Manag. Res.*, 12, 507-526.

Van der Voet, E. 2002. Substance flow analysis methodology, in Ayres, R.U., Ayres, L.W. (Eds.), *A Handbook of Industrial Ecology*, Edward Elgar Publishing Ltd., Cheltenham, UK, pp.91-101.

Viavattene, C., Revitt, M., Scholes, L., 2010. Cost implications of different source control treatment options: industrial treatment and storm water BMP. Deliverable 8.5b of the ScorePP project.
<http://documents.er.dtu.dk/Projects/ScorePP/ScorePP%20D8.4%20Socio-economic%20benchmarks,%202010-05-14.pdf> (last accessed 1 March 2012)

Visesmanee, V. Polprasert, C. and Parkpian, P. 2008. Long-term performance of subsurface-flow constructed wetlands treating Cd wastewater. *J Environ Sci Health-Part*, 43, 765-771.

Wu, J., Y.S. Yang, Y. S. and Lin, J. (2005). Advanced tertiary treatment of municipal wastewater using raw and modified diatomite. *J Hazard Mater*, B127, 196–203.